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Exciton Spin States in Nanocrystal Quantum Dots Revealed by Spin-Polarized Resonant PL and Raman Spectroscopy

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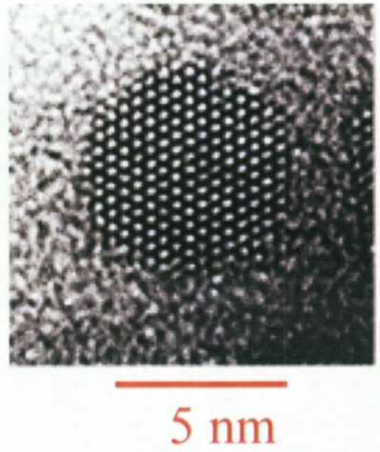
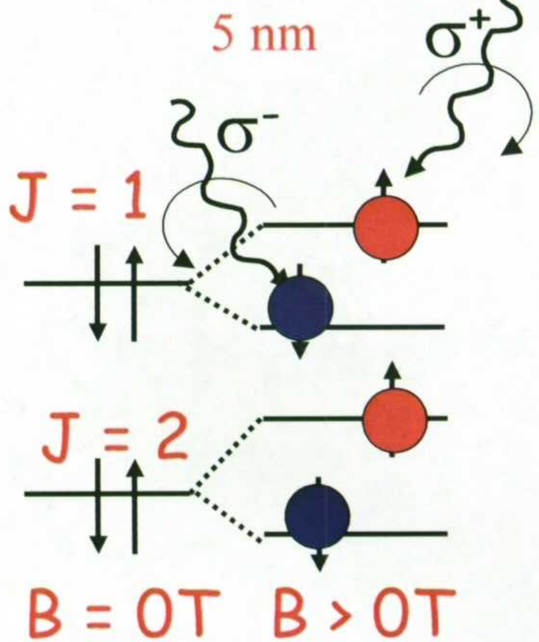
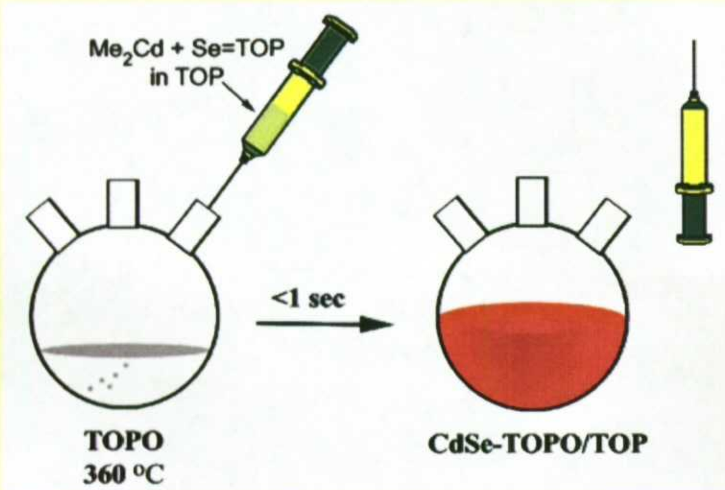
Outline

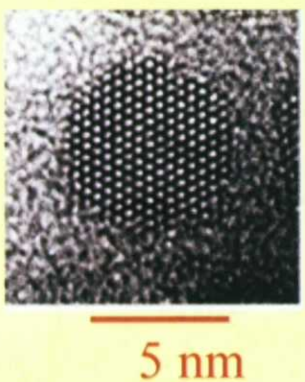
- ❑Brief introduction to nanocrystal quantum dots
- ❑Fluorescence line narrowing, “dark” and “bright” excitons in colloidal CdSe nanocrystal quantum dots
- ❑ Polarization -resolved high-resolution resonant photoluminescence measurements in high magnetic fields: probing the exciton spin states
- ❑ Electron-hole exchange interaction and the exciton Zeeman splitting in CdSe nanocrystals
- ❑Conclusions

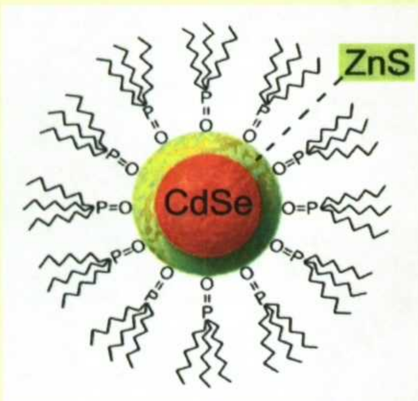




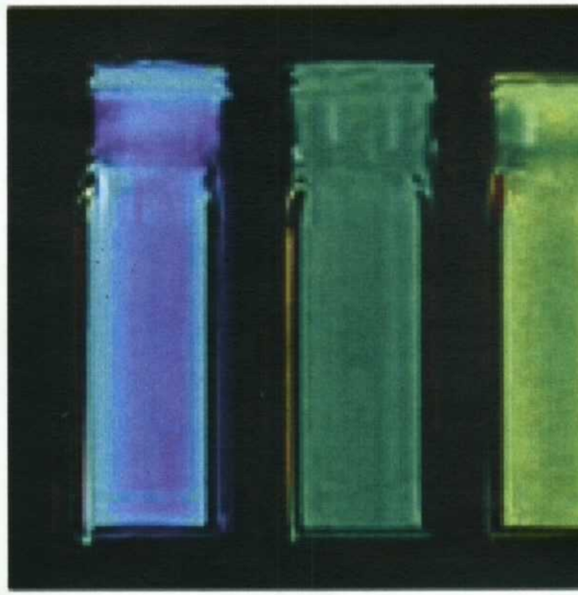
Colloidal CdSe Nanocrystal Quantum Dots







Size-tunable optical properties

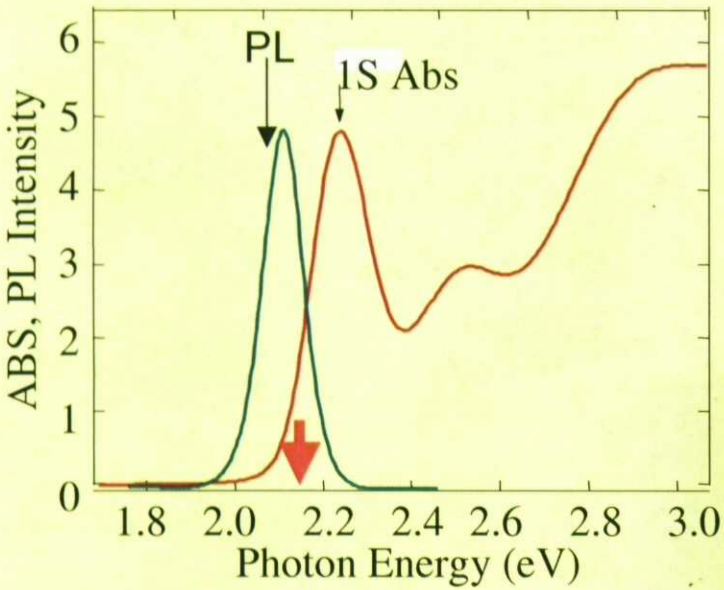


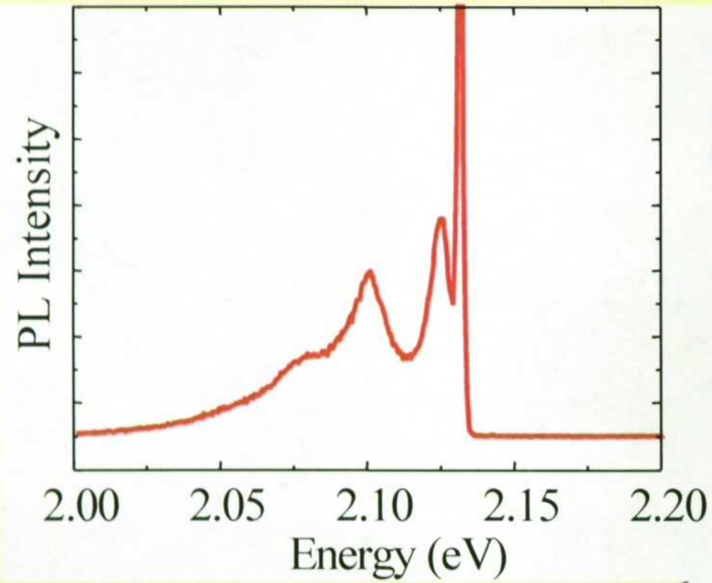
0.9nm 1.4nm 1.9nm 2.4nm

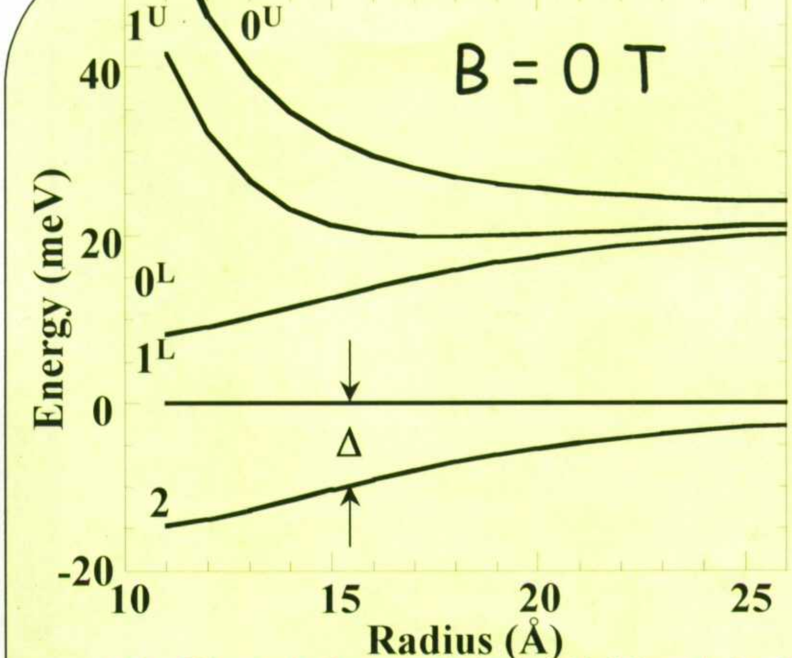
Average radius: 1nm~10nm

Size dispersion: 5%

Fluorescence Line Narrowing (Resonant Excitation)





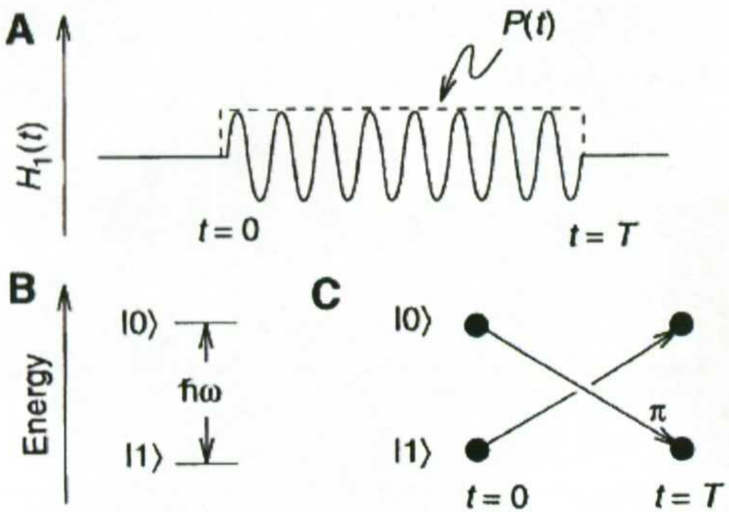


A. L. Efros, M. Rosen, M. Kuno, M. Nirmal, D. J. Norris, and M. Bawendi, *Phys. Rev. B* **54**, 4843 (1996).

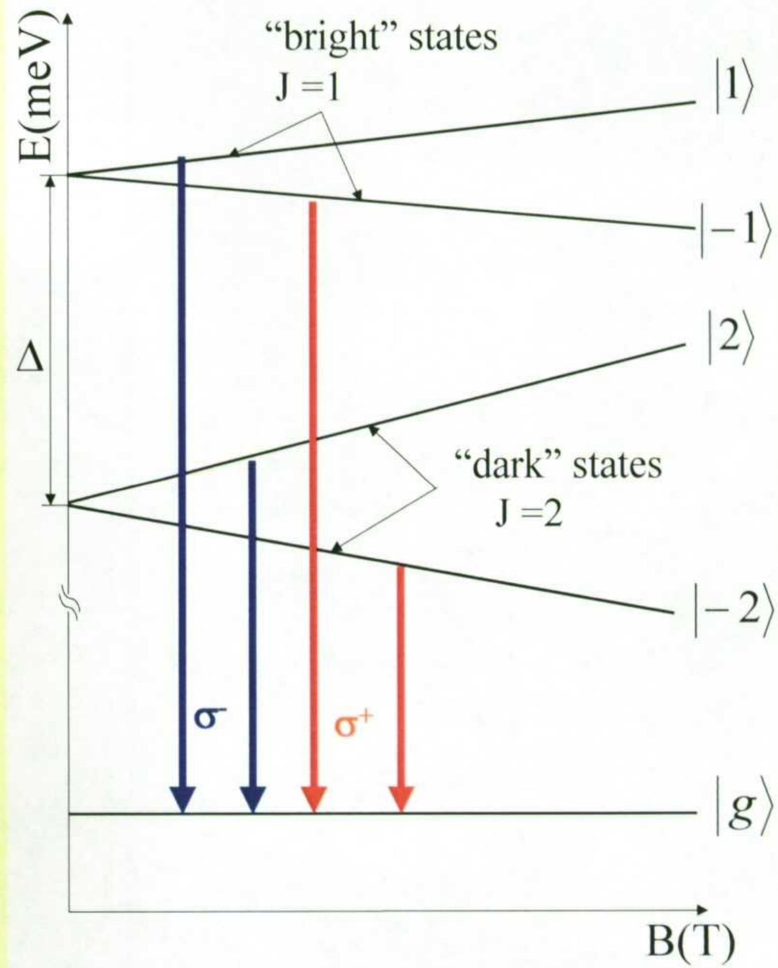
- Electron-hole interaction and the wurtzite symmetry lift the eight-fold degeneracy of the exciton ground state
- The result is a five-level "exciton fine structure"

Probing Exciton Spin States in Quantum Dots

One qubit "NOT" gate

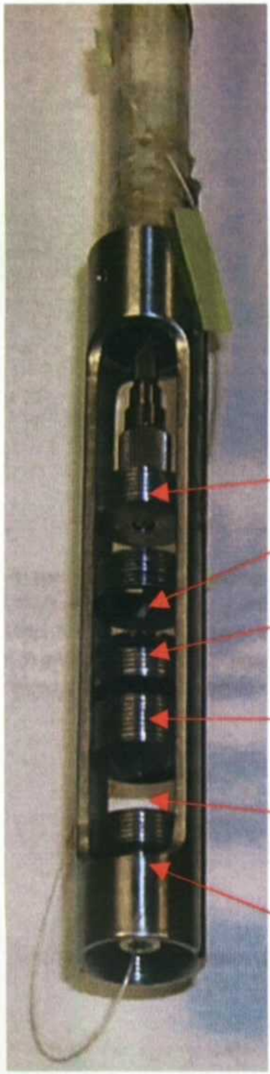


D. P. DiVincenzo, Science 270, 255 (1995).



Experiment

specially designed fiber-coupled probe



collection fiber

sample mount

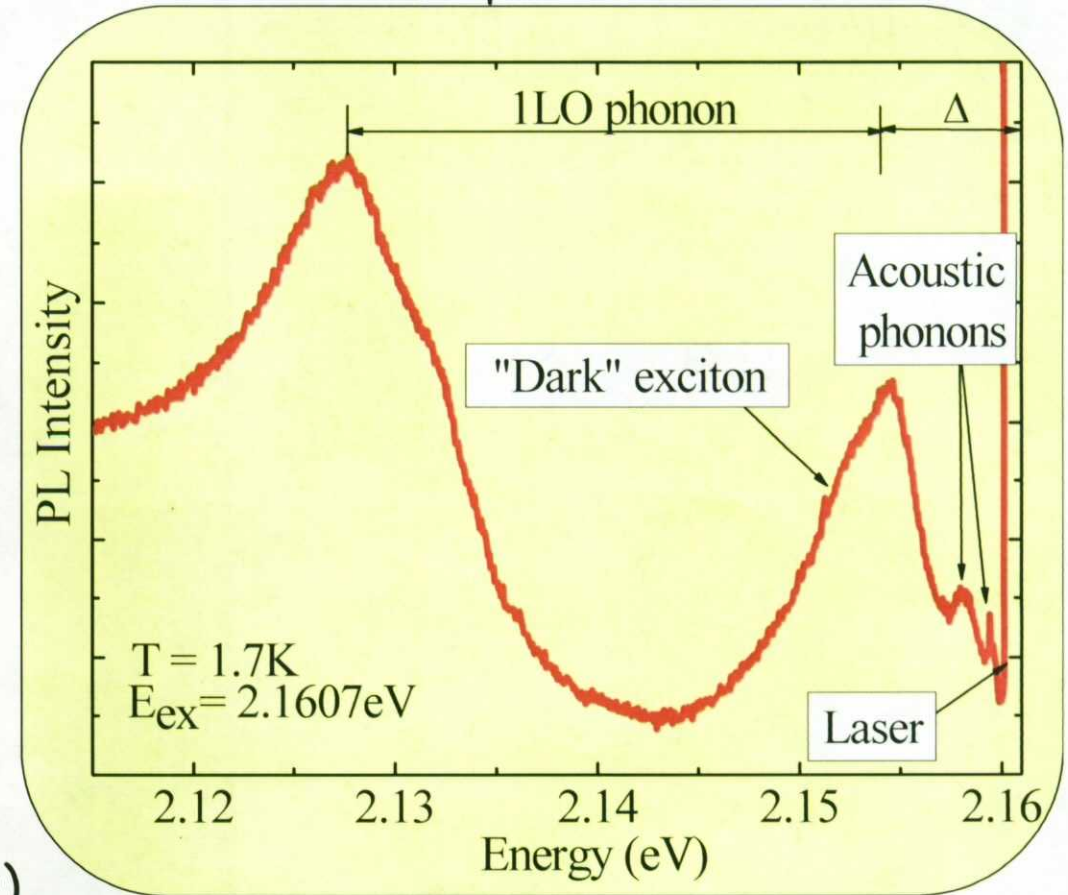
parabolic reflector

focusing lens

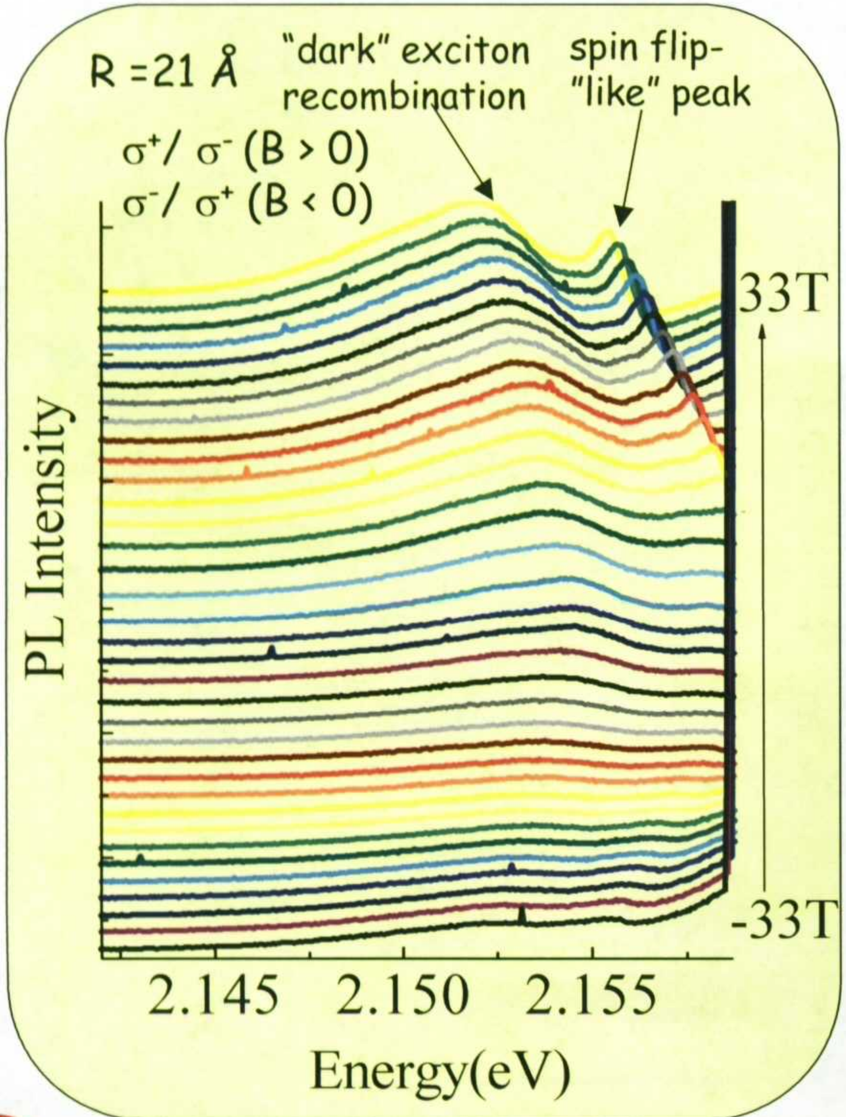
circular polarizer

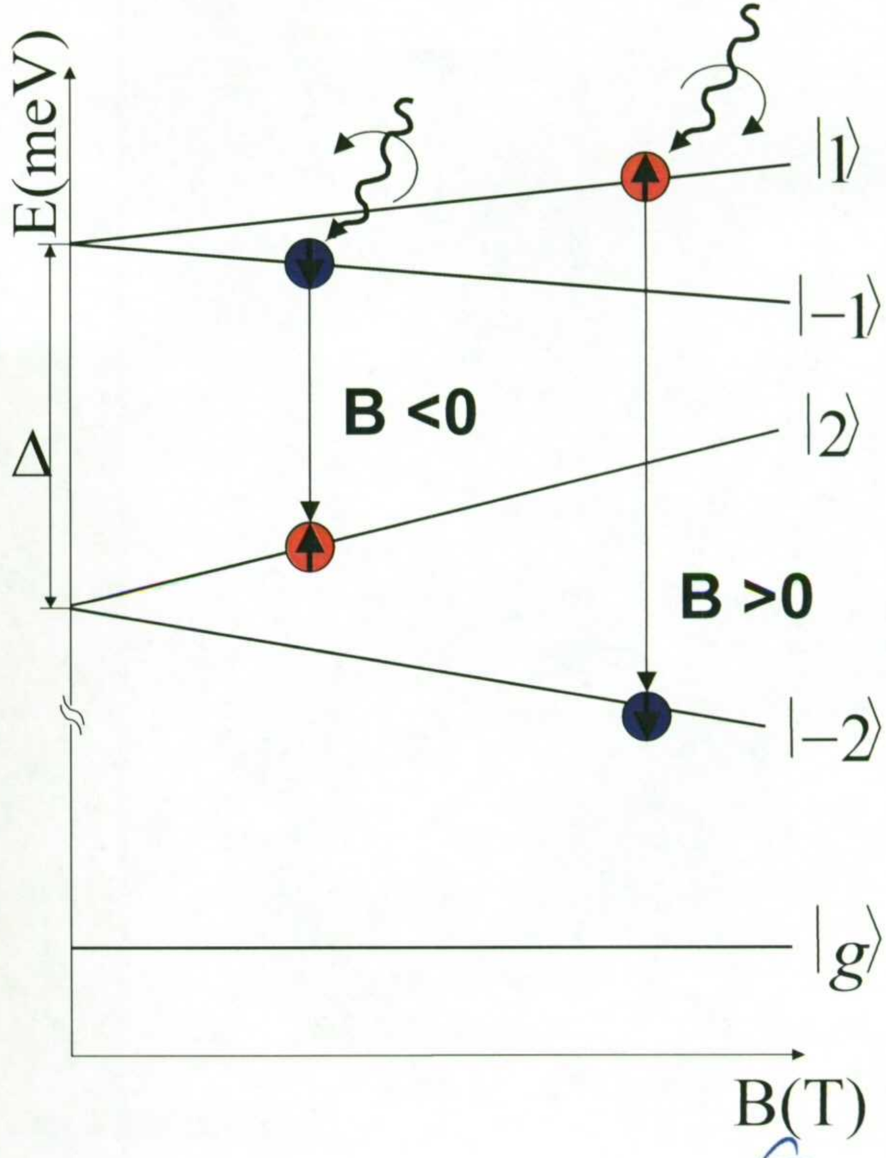
delivery fiber (from tunable laser)

Zero-field high resolution resonant PL spectrum

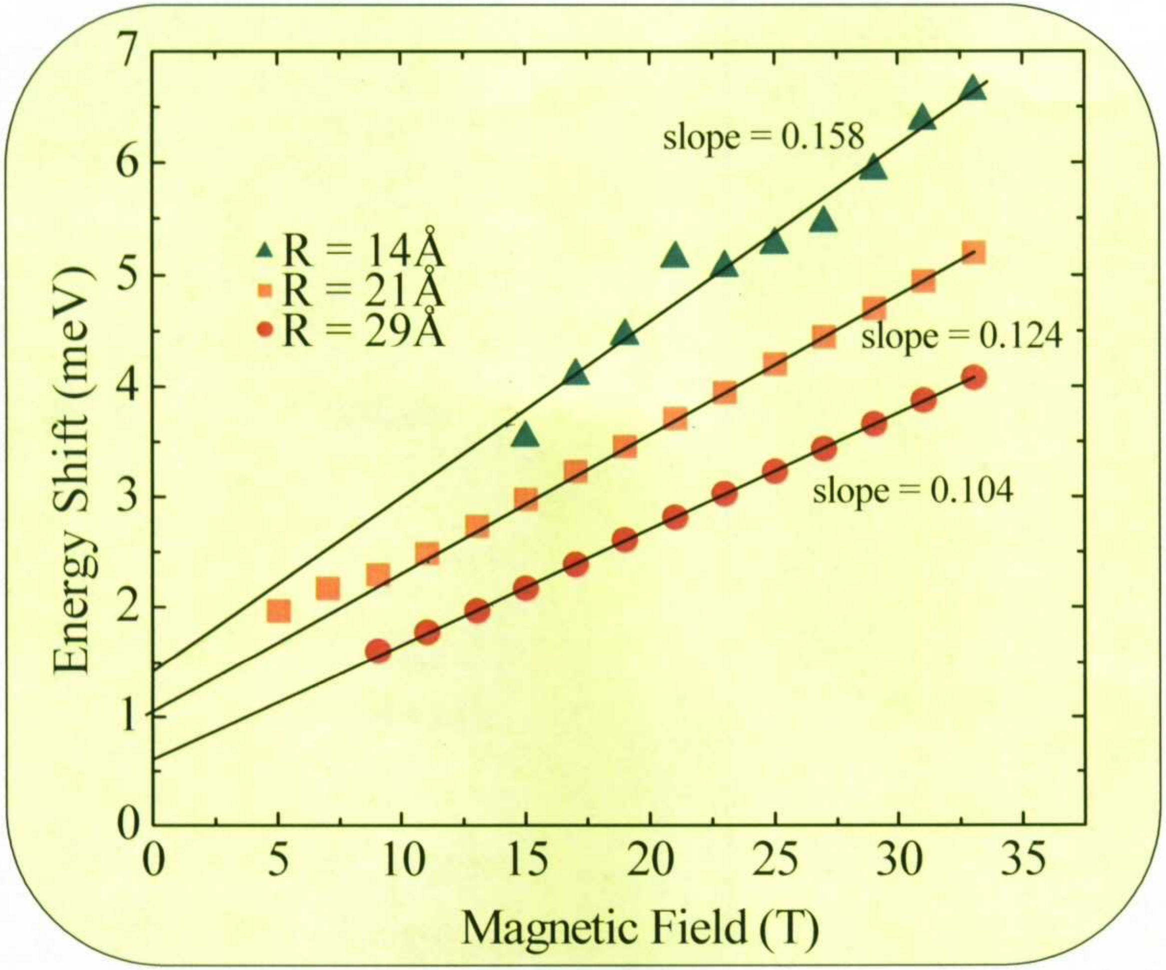


PL Spectra as a Function of Magnetic Field





The "Spin Flip-Like" Transition



Extrapolation of the linear fit at 0T is finite!- peak cannot be associated with free electron spin flip.

Electron-Hole Exchange Interaction Hamiltonian in a Magnetic Field

$$
\begin{array}{c|cccc}
 & |-2\rangle & |-1\rangle & |1\rangle & |2\rangle \\
\hline
|-2\rangle & -\frac{1}{2}g_2\mu_B B_z & \frac{1}{2}g_e\mu_B B_x & 0 & 0 \\
|-1\rangle & \frac{1}{2}g_e\mu_B B_x & \Delta-\frac{1}{2}g_1\mu_B B_z & \frac{1}{2}\alpha & 0 \\
|1\rangle & 0 & \frac{1}{2}\alpha & \Delta+\frac{1}{2}g_1\mu_B B_z & \frac{1}{2}g_e\mu_B B_x \\
|2\rangle & 0 & 0 & \frac{1}{2}g_e\mu_B B_x & \frac{1}{2}g_2\mu_B B_z
\end{array}
$$

$$g_1 = -g_e + 3g_h$$

$$g_2 = g_e + 3g_h$$

g_e, g_h = electron, hole g-factors

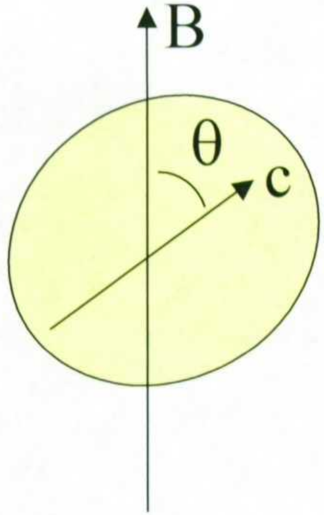
z-direction = c-axis

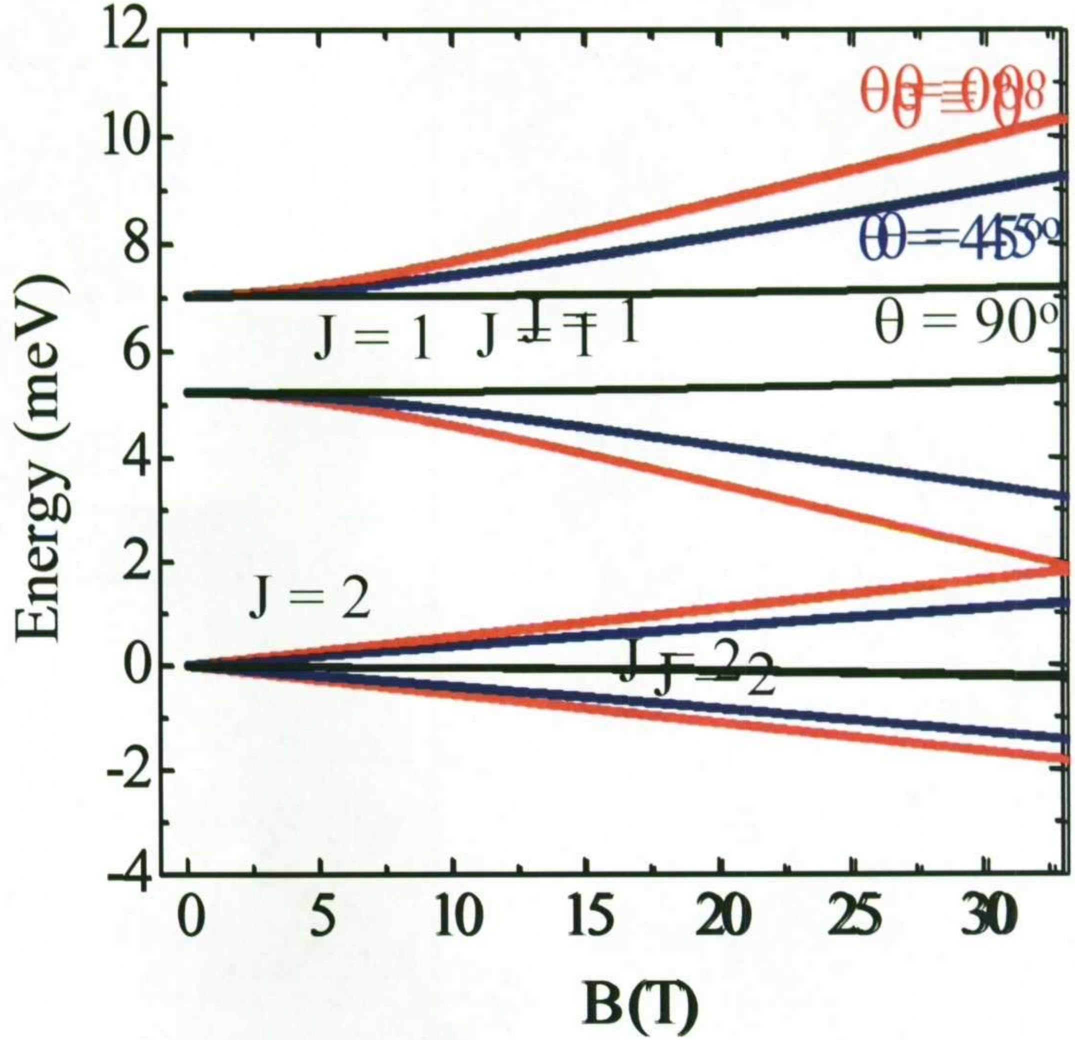
Δ = "dark"-"bright" splitting

α = zero field splitting of the J=1 state

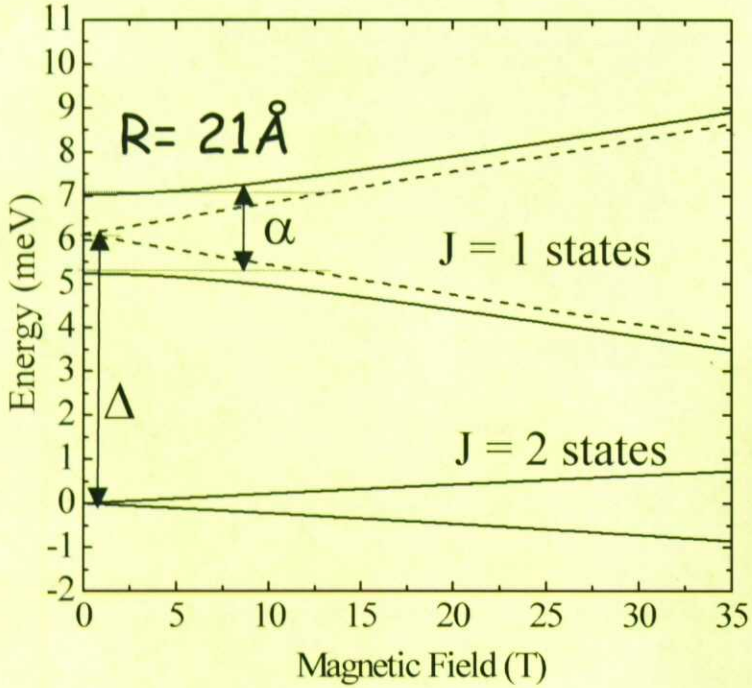
Magneto-PL studies of self-assembled CdSe/ZnSe QDs-long range exchange interaction is responsible for a zero-field Zeeman splitting of the J = 1 state (J. Puls, M. Rabe, H. -J. Wünsche, and F. Henneberger, *Phys. Rev. B* **60**, R-16303 (1999)).

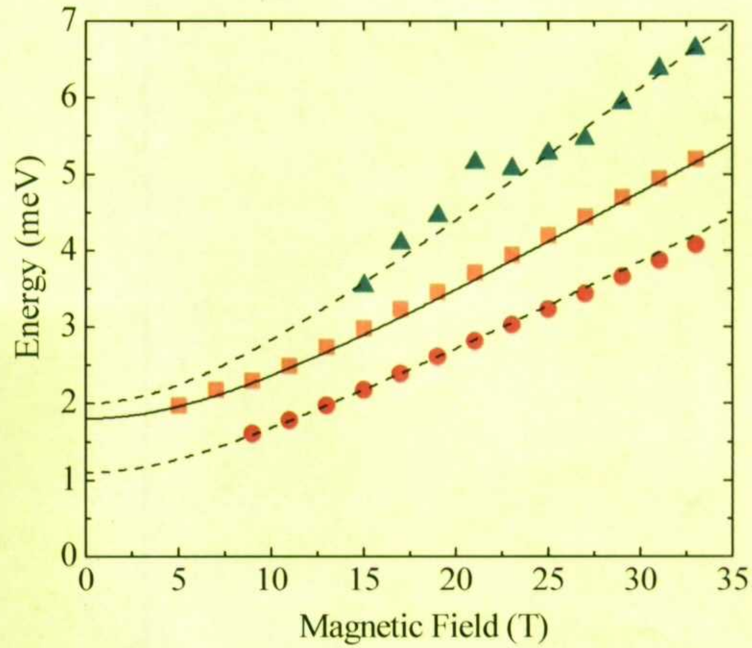
Zeeman Splitting and the Crystal Orientation





Zeeman Splitting of the J=1 State





R(Å)	Δ^{**}(meV)	α(meV)	g_e^{*}	g_h
14	13.2	2	1.6	-1
21	6.13	1.8	1.2	-0.8
29	3	1.1	0.8	-0.7

*spin precession measurements J. A. Gupta, D. D. Awschalom, Al. L. Efros, and A. V. Rodina, *Phys. Rev. B* **66**, 125307 (2002).

**measured in the present experiment

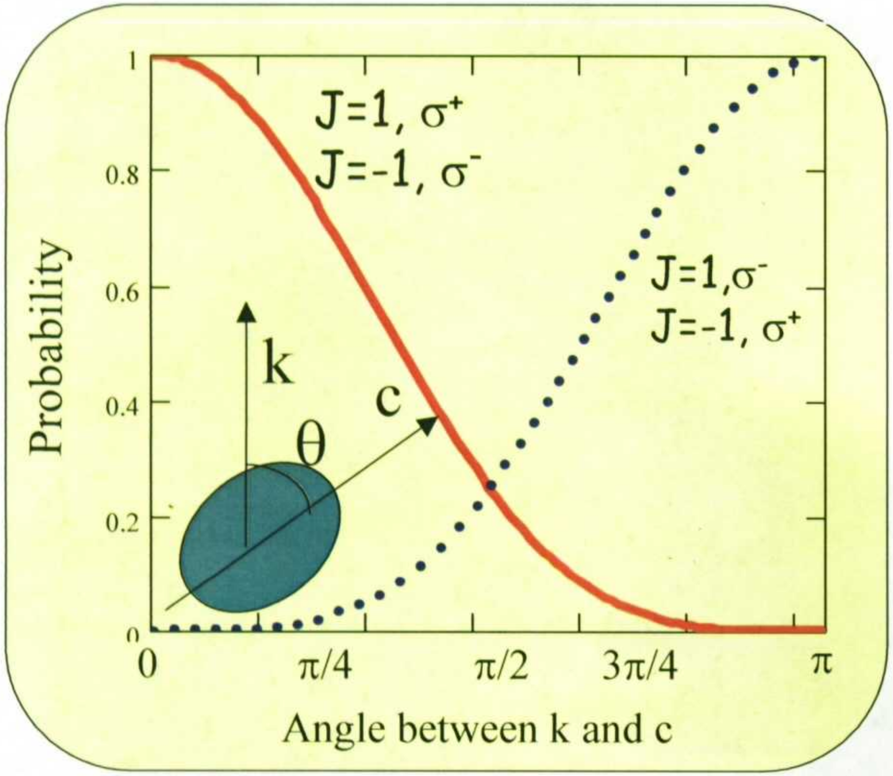
Conclusions

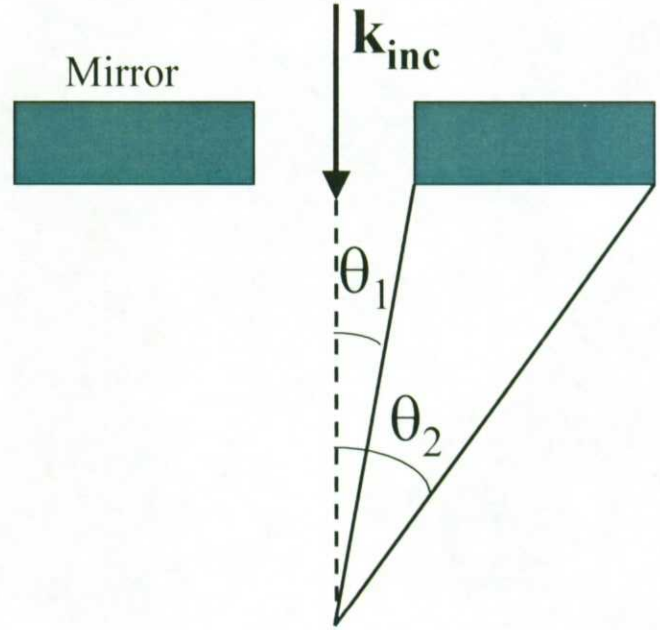
- Exciton spin states were probed by spin-resolved high resolution magneto-PL experiments
- Developed fiber-coupled probe that minimizes collection of scattered excitation
- Observed transition associated with the spin-flip of J = 1 excitons
- Finite splitting at OT –long range exchange interaction?
- Raman spin-flip or photoluminescence?

Acknowledgements

A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by NSF Cooperative Agreement No. DMR-0084173, by the State of Florida, and by the DOE

Zeeman Splitting for Randomly Oriented QD





$$\langle E(B)\rangle = E(0) + \frac{1}{4\pi}\int_0^{2\pi}\int_{\theta_1}^{\theta_2}(E(\theta,B)-E(0))\frac{1}{4}(1+\cos(\theta))^2\sin(\theta)\,d\theta\, d\varphi$$

$$E(0) = 0 \ \ for\ J = 2\ states$$

$$E(0) = \Delta - \delta/2 \ \ for\ J = -1\ state$$

$$E(0) = \Delta + \delta/2 \ \ for\ J = 1\ state$$